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AASERT-93 NEW HIGH-PRESSURE DIAGNOSTIC TECHNIQUE

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11. SUPPLEMENTARY NOTES

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13. ABSTRACT (Maximum 200 words)

As we outlined in our submi-<sup>ted</sup> proposal, progress towards making DFWM spectroscopy quantitative has been achieved. This work makes explicit how the magnitude of the DFWM signal depends on the polarizations of the three incident beams under the weak and strong-field limits. We have been using DFWM to investigate acetylene (C2H2) and methyl radical (CH3) molecules in an atmospheric pressure flame and in a low-pressure hot-filament reactor. To calibrate the measurement, acetylene is measured in the free flow of a C2H2/O2 mixture, and also in the pre-reaction zone of a C2H2/O2 flame -- both with a fast flow rate of 40-50 m/s at the nozzle outlet of the mixture. The DFWM signal falls in the weak field limit (far from saturation), which means the DFWM signal is proportional to the products of the three incident beam intensities (If1b1p).

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**TECHNICAL REPORT**  
**Progress in Developing New Diagnostic Technique**  
**08/01/93 - 07/31/94**

Degenerate Four-Wave Mixing (DFWM) is a nonlinear optical process that can be used as a nonintrusive *in situ* diagnostic of hostile environments such as plasmas, flames, arcs and explosions. By identifying molecular species and measuring their concentration, temperature and internal-state population, we can understand better complex chemical environments and mechanisms.

As we outlined in our submitted proposal, progress towards making DFWM spectroscopy quantitative has been achieved [1][2] (mainly by Dr. Skip Williams, who graduated in January, 1994 and who is a post-doctoral fellow at Los Alamos National Laboratory). This work makes explicit how the magnitude of the DFWM signal depends on the polarizations of the three incident beams under the weak and strong-field limits.

We have been using DFWM to investigate acetylene ( $C_2H_2$ ) and methyl radical ( $CH_3$ ) molecules in an atmospheric pressure flame and in a low-pressure hot-filament reactor [3] in collaboration with Professor C. Kruger, Department of Mechanical Engineering, Stanford University. We have measured DFWM spectra of the  $\tilde{A}^1A_u - \tilde{X}^1\Sigma_g^+$  transition in acetylene at wavelengths around 216 nm. To calibrate the measurement, acetylene is measured in the free flow of a  $C_2H_2/O_2$  mixture, and also in the pre-reaction zone of a  $C_2H_2/O_2$  flame --- both with a fast flow rate of 40-50 m/s at the nozzle outlet of the mixture. The DFWM signal falls in the weak field limit (far from saturation), which means the DFWM signal is proportional to the products of the three incident beam intensities ( $I_f I_b I_p$ ). When the reactor is filled with 1 atm pressure of acetylene, only an unknown DFWM spectrum is recorded, which we speculate is from the products of acetylene dissociation because acetylene is not transported to the detection zone as efficiently as in the fast flow. Similar results occur for the DFWM of the methyl radical. Molecules like electronically excited acetylene or methyl radical are metastable species that predissociate; for DFWM to succeed the only alternative is to avoid the electronic excitation region of the above species and shift to the vibrational excitation region in the near IR. This shift needs a totally new laser system to initiate it.

A new optical diagnostic: cavity ring-down spectroscopy [4] is another possibility to overcome the above problems at 216nm; by this means methyl radicals have been detected and more detailed measurements are in progress.

The AASERT student's grades are satisfactory

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## References

1. S. Williams, R.N. Zare and L.A. Rahn, "Reduction of Degenerate Four-Wave Mixing Spectra to relative Populations I: Weak-Field Limit," J. Chem. Phys. **101**, 1072 (1994)
2. S. Williams, R.N. Zare and L.A. Rahn, "Reduction of Degenerate Four-Wave Mixing Spectra to relative Populations II: Strong-Field Limit," J. Chem. Phys. **101**, 1093 (1994)
3. M.L. Menningen, etc. Chem. Phys. Lett. **204**, 573 (1993)
4. A.O'Keefe and D.A.G. Deacon, Rev. Sci. Instrum. **59**, 2544 (1988)

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